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Ordered nanocolumn-array organic semiconductor thin films with controllable molecular orientation

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ABSTRACT

Ordered nanocolumn-array phthalocynine semiconductor thin films with controllable molecular orientation were fabricated by combining molecular template growth (MTG) and glancing angle deposition (GLAD) techniques. The pre-deposited planar perylene-3,4,9,10-tetracarboxylic-3,4,9,10-dianhydride (PTCDA) molecular template layer induces phthalocynine molecules arrange with a lying-down molecular orientation, in which the π–π stacking is vertical to the substrate improving the charge transport along the vertical direction; While the GLAD technique supports the formation of nanocolumn-array thin films, supplying a much larger exposed surface area than the conventional compact thin films. The ordered nanocolumn-array thin films with controllable molecular orientation fabricated by combining MTG and GLAD techniques show the potentials to fabricate ordered bulk heterojunction for improving the performance in organic photovoltaics.

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1. Introduction

Compared with conventional inorganic electronics, organic electronics are lighter, more flexible, and less expensive. Particularly, organic electronics can be processed in large area with high output by roll-to-roll printing techniques [1–4]. Accordingly, organic electronics shows a desirable alternative in many applications as well as creates the possibility of new applications that would be impossible using conventional inorganic electronic materials, such as organic field effect transistors (OFETs), organic photovoltaics (OPVs), organic light-emitting diodes (OLEDs), smart windows, electronic paper [5–8]. Organic active layer thin film is one of the key components in organic electronic devices. Its quality directly determines the device performance. Lots of studies have shown that the performance of organic electronic devices could be improved through optimizing the organic active layer morphology and structure [9–13]. Accordingly, it is of great importance to develop techniques to fabricate high-quality organic active layer thin films with controllable morphology and structure, for example, patterning, photolithography, molecular template growth (MTG), glancing angle deposition (GLAD) [14–20].

The MTG was developed recently to fabricate high-quality organic semiconductor thin films for improving the performance of OFETs and OPVs respectively using different molecular template layer materials [12,17,21]. For example, the *para*-sexiphenyl (*p*-6P) as a molecular template layer could support the formation of single-crystal-like thin films because of the epitaxial relationship between *p*-6P and the overlayer crystals, dramatically improving the charge mobilities [12]; While the perylene-3,4,9,10-tetracarboxylic-3,4,9,10-dianhydride (PTCDA) molecular template layer induce overlayer molecules with a lying-down arrangement, improving the charge transport along the vertical direction and accordingly improving the OPV performance [17].

The GLAD, as one of vacuum deposition techniques, has been of interest to control three-dimensional (3-D) nanoscale structured thin films for nearly 20 years [22–24]. It is an *in-situ* sculpting technique to create 3-D nano-structured thin film such as pillars, columns, helixes, zig-zags, etc. through shadowing effects. The nanostructured thin films can be controlled by the deposition rate, the substrate temperature, the substrate rotate rate as well as the glancing angle. At early stages, GLAD was mainly used to fabricate metal and inorganic semiconductor thin films with controllable 3-D nanoscale structures [22]. Recently it was also successfully developed to fabricate organic semiconductor thin films with nanocolumns, and showed the potential application in OPVs [19,20,25–28].

In our previous report, PTCDA molecular template growth has already been used to grow organic semiconductor thin films with controllable molecular orientation, and the performance of planar heterojunction OPVs based on PTCDA template growth was improved [17]. However, the limited donor–acceptor interface in planar heterojunction hinders the effective formation and

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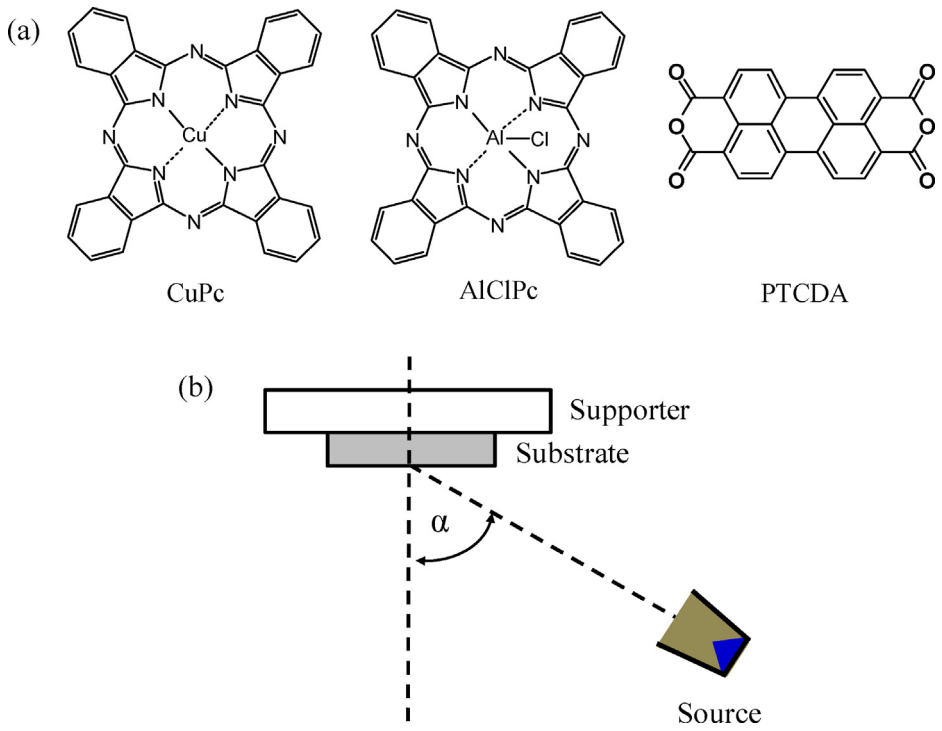

Fig. 1. (a) Molecular structures of CuPc, AlClPc and PTCDA. (b) Schematic of glancing angle deposition (GLAD). The glancing angle was set at 65° (normally $\alpha < 30°$ in the conventional OMBD).

separation of the excitons. It is desirable to develop methods to fabricate ordered bulk heterojunction for improving the exciton formation and separation as well as the charge transport and collection [29]. In present paper, we combine MTG and GLAD techniques to fabricate ordered nanocolumn-array phthalocynine semiconductor thin films with a controllable lying-down molecular orientation in the models of planar molecule copper phthalocyanine (CuPc) and non-planar molecule chloroaluminum phthalocyanine (AlClPc). The nanocolumn-array thin films with lying-down molecular orientation show great potentials to form ordered bulk heterojunction for improving the performance in OPVs.

2. Experimental

The PTCDA (97%), CuPc (99%), and AlClPc (85%) materials (Fig. 1a) were purchased from Sigma-Aldrich and purified twice by thermal gradient sublimation prior to use. The high-vacuum organic molecular beam deposition (OMBD) at a base pressure of $\sim 6 \times 10^{-4}$ Pa was used to fabricate organic semiconductor thin films at ambient temperature. All of the films were grown on commercially available SiO_2 or ITO substrates with a root-mean-square roughness (RMS) of about 0.4 nm and 1.0 nm, respectively. The PTCDA template layers with thicknesses of 1 nm and 5 nm were first deposited on the SiO_2 or ITO substrates using conventional OMBD followed by deposition of the CuPc and AlClPc layer with the GLAD technique (Fig. 1b), in which the glancing angle α was set at 65° (normally the glancing angle can be controlled between 30° and 90°, and $\alpha < 30°$ in the conventional OMBD) without rotation. The film thickness and growth rate were monitored by calibrated quartz microbalances. The deposition rates of 0.15–0.20 Å/s were used for the materials. Reference samples of PTCDA, CuPc, AlClPc films were also grown on SiO_2 or ITO for direct comparison with the MTG and GLAD samples as well as the samples fabricated by combining MTG and GLAD.

The morphology of thin films was characterized by tapping-mode atomic force microscopy (Agilent Technologies 5500 AFM/SPM System, USA) and field-emission scanning electron microscopy (FE-SEM, Nova NanoSEM 230, FEI Electron Optics B.V,

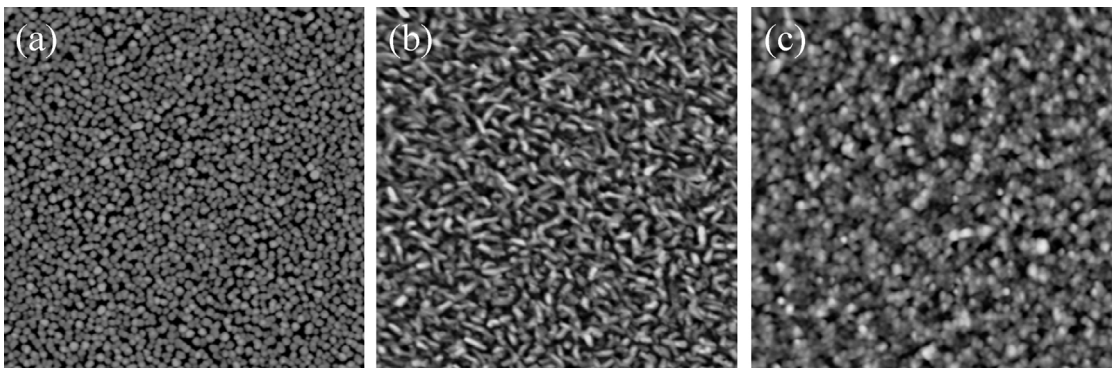

Fig. 2. The 1.0 μm × 1.0 μm AFM images of PTCDA and CuPc thin films fabricated by conventional OMBD. (a) 1 nm PTCDA/SiO_2 (RMS ≈ 1.0 nm), (b) 50 nm CuPc/SiO_2 (RMS ≈ 2.2 nm), (c) 50 nm CuPc/1 nm PTCDA/SiO_2 (RMS ≈ 4.5 nm).

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